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On a Connection Between Nonlinear Response to an External Field and Equilibrium Properties of Systems with Interaction: Comment on cond-mat/9711226 by Kopietz and Völker

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We demonstrate the danger of a naive identification of kinetic coefficients in the free-electron response expansion in powers of an *external* electric field and coefficients in the expansion of the corresponding equilibrium quantity in powers of the fluctuating *intrinsic* electric field. A particular example of the direct current generation by an AC electric field in a mesoscopic system of non-interacting electrons vs. the Fock contribution to the equilibrium persistent current in a system of interacting electrons is considered. We comment on a recent paper by Kopietz and Völker, cond-mat/9711226.

The problem of an adequate theoretical description of electron-electron interaction and electron relaxation processes in disordered mesoscopic conductors has become a very hot topic nowadays [1]. The complexity of the problem stimulates the search for approaches which incorporate general relationships between kinetic and equilibrium properties of the system. One may believe that the knowledge of the nonlinear response to an external field supplemented by the field correlation functions obtained, e.g., with the use of the fluctuation-dissipation theorem (FTD) [2] is helpful for solving the problem.

The purpose of this Letter is to demonstrate how dangerous might be a naive way of applying FDT-like relations for description of systems of *interacting* electrons, when the coefficients of expansion of an equilibrium quantity in powers of the fluctuating intrinsic electric field are identified with the kinetic coefficients in the free-electron response expansion in powers of an external electric field.

The reason is a deep physical difference between the dissipative nature of the causal response to an external field and the "non-causal" nature of all the interaction processes in the equilibrium.

As an actual example we consider the nonlinear generation of the direct current $I_{DC}^{(2)}$ of *non-interacting* electrons in a mesoscopic ring of the circumference L pierced by a constant magnetic flux ϕ and an AC flux ϕ_ω that produces an electric field $\mathcal{E}(\omega) = (i\omega/Lc)\phi_\omega$. This kinetic problem has been studied earlier in Ref. [3], [4] and an expression for the nonlinear conductance $\sigma_{kin}^{(2)}(\omega, -\omega)$ defined by

$$I_{DC}^{(2)} = \sigma_{kin}^{(2)}(\omega, -\omega)|\mathcal{E}_\omega|^2, \tag{1}$$

has been obtained. A peculiar feature of the result of Refs. [3], [4] is that the nonlinear conductance $\sigma_{kin}^{(2)}(\omega, -\omega)$ does not vanish exponentially in the limit of high frequencies $\omega \gg E_c$, where $E_c = D/L^2$ is the Thouless energy and D is the diffusion coefficient. This is in a striking contrast to a similar mesoscopic effect of the Aharonov-Bohm oscillations of the linear conductance [5].

Recently there was an attempt by Kopietz and Völker [6] to relate the nonlinear conductance $\sigma_{kin}^{(2)}(\omega, -\omega)$ with the coefficient $\sigma_{eq}^{(2)}(\mathbf{q}, i\omega_n)$ in the expression for the Fock (exchange) contribution to the equilibrium persistent current in the system of *interacting* electrons:

$$I_{PC}^{(F)} = -\frac{T}{2}\sum_{\mathbf{q},n} \sigma_{eq}^{(2)}(\mathbf{q}, i\omega_n) \frac{\langle (\mathbf{q}\cdot\mathbf{E}_{\mathbf{q}}(i\omega_n))(\mathbf{q}\cdot\mathbf{E}_{-\mathbf{q}}(-i\omega_n))\rangle}{|\mathbf{q}|^2}. \tag{2}$$

Here ω_n are bosonic Matsubara frequencies $\omega_n = 2\pi T n$, and $\langle E_{\mathbf{q}}^{\alpha}(i\omega_n)\, E_{-\mathbf{q}}^{\beta}(-i\omega_n)\rangle = q_\alpha q_\beta\, D(\mathbf{q}, i\omega_n)$ is proportional

to a disorder-averaged Green's function of longitudinal photons [7] $D(\mathbf{q},i\omega_n)=4\pi/q^2\varepsilon(\mathbf{q},i|\omega_n|)$ in a media with the dielectric function $\varepsilon(\mathbf{q},\omega)$.

One can transform Eq.(2) to an integral over real frequencies by representing the sum in terms of a contour integral.

$$I^{(F)}_{PC}=I^{st}_{PC}+\sum_{\mathbf{q}}\int_0^{+\infty}\frac{d\omega}{2\pi}\,I^{(F)}_{PC}(\mathbf{q},\omega), \tag{3}$$

where I^{st}_{PC} is a "static" contribution given by the $\omega_n=0$ term of Eq.(2) and

$$I^{(F)}_{PC}(\mathbf{q},\omega)=\coth\left(\frac{\omega}{2T}\right)\,\Re\left[i\sigma^{(2)}_{eq}(\mathbf{q},\omega)\,q^2D(\mathbf{q},\omega)\right]. \tag{4}$$

Here $\sigma^{(2)}_{eq}(\mathbf{q},\omega)=[\sigma^{(2)}_{eq}(-\mathbf{q},-\omega)]^*$ and $D(\mathbf{q},\omega)=[D(-\mathbf{q},-\omega)]^*$ are analytical continuations of $\sigma^{(2)}_{eq}(\mathbf{q},i\omega_n)$ and $D(\mathbf{q},i\omega_n)$ in the upper half-plane of ω.

Eq.(4) can be further simplified in the Ohmic regime characterized by a constant conductivity σ_0, where $\varepsilon(\mathbf{q},\omega)=4\pi\sigma_0/(-i\omega)$. In this case $D(\mathbf{q},\omega)$ is purely imaginary and is related with the spectral density of the fluctuating intrinsic electric filed $\langle|\mathbf{E}_{\mathbf{q},\omega}|^2\rangle$ by the FDT [2]:

$$\langle|\mathbf{E}_{\mathbf{q},\omega}|^2\rangle=i\coth\left(\frac{\omega}{2T}\right)\,q^2D(\mathbf{q},\omega)=\coth\left(\frac{\omega}{2T}\right)\,\frac{\omega}{\sigma_0}. \tag{5}$$

Thus we arrive at an expression analogous to Eq.(1):

$$I^{(F)}_{PC}(\mathbf{q},\omega)=\Re\,\sigma^{(2)}_{eq}(\mathbf{q},\omega)\,\langle|\mathbf{E}_{\mathbf{q},\omega}|^2\rangle. \tag{6}$$

It is tempting to interpret Eq.(6) as a "nonlinear response" to the fluctuating intrinsic field $\mathbf{E}_{\mathbf{q},\omega}$ in the system of interacting electrons, by an analogy with the response Eq.(1) to an external field $\mathcal{E}_{\mathbf{q},\omega}$ applied to a free electron system.

Then one may believe [6] that there is a simple relationship between $\sigma^{(2)}_{kin}(\omega,-\omega)$ in Eq.(1) and $\sigma^{(2)}_{eq}(\mathbf{q},\omega)$ in Eq.(6):

$$\sigma^{(2)}_{kin}(\omega,-\omega)=\lim_{\mathbf{q}\to 0}\,\Re\sigma^{(2)}_{eq}(\mathbf{q};\omega). \tag{7}$$

However, this leads to a dramatic discrepancy with Ref. [3,4], since $\sigma^{(2)}_{eq}(\mathbf{q};\omega)$ appears to be [6] exponentially small at $\omega\gg E_c$, while the $\sigma^{(2)}_{kin}(\omega,-\omega)$ decreases only as ω^{-2} [3,4].

Below we will show that the relationship (7) is *principally wrong*. This means that one cannot obtain a correct Fock contribution to the equilibrium persistent current by substituting $\Re\sigma^{(2)}_{eq}(\mathbf{q};\omega)$ for the free-electron nonlinear conductance $\sigma^{(2)}_{kin}(\mathbf{q},\omega,-\omega)$ (at a finite momentum $\mathbf{q}$) into Eq.(6).

To this end we use the expression [6] for $\sigma^{(2)}_{eq}(\mathbf{q},i\omega_n)$ as the derivative of the free-electron polarization operator $\Pi_0(\mathbf{q},i\omega_n)$ over the magnetic flux ϕ:

$$\sigma^{(2)}(\mathbf{q},i\omega_n)=\frac{ce^2}{2q^2}\frac{\partial}{\partial\phi}\overline{\Pi_0(\mathbf{q},i\omega_n)}, \tag{8}$$

where the overline denotes the disorder average and

$$\Pi_0(\mathbf{q},i\omega_n)=T\sum_{\mathbf{k},\mathbf{k'},l}G_{\mathbf{k}+\mathbf{q},\mathbf{k'}+\mathbf{q}}(i\epsilon_l+i\omega_n)G_{\mathbf{k'},\mathbf{k}}(i\epsilon_l)\equiv T\sum_l Tr_{\mathbf{k}}G(i\epsilon_l+i\omega_n)\,G(i\epsilon_l). \tag{9}$$

Here G are Matsubara Green's functions of non-interacting electrons in the presence of disorder; $\epsilon_l=(2l+1)\pi T$, and the operation $Tr_{\mathbf{k}}$ is defined to shorten the following expressions.

The analytical continuation in Eq.(9) can be performed in a general form:

$$\begin{aligned}\Pi_0(\mathbf{q},\omega)=Tr_{\mathbf{k}}\int_{-\infty}^{\infty}\frac{d\epsilon}{4\pi i}\,&\Big[[G^R(\epsilon+\omega)G^R(\epsilon)-G^A(\epsilon)G^A(\epsilon-\omega)]\tanh\left(\frac{\epsilon}{2T}\right)+\\&+2G^R(\epsilon+\omega)G^A(\epsilon)\,[n_F(\epsilon)-n_F(\epsilon+\omega)]\Big],\end{aligned} \tag{10}$$

where $n_F(\epsilon)=[\exp{(\epsilon/T)}+1]^{-1}$ and the retarded (R) and advanced (A) electron Green's functions are the analytical continuations of the Matsubara Green's function $G(\epsilon)$ into the upper $(\mathrm{Im}\,\epsilon>0)$ and lower $(\mathrm{Im}\,\epsilon<0)$ half-planes, respectively.

The flux derivative corresponds to breaking of one Green's function into two by the current vertex $\hat{j}$:

$$\frac{\partial}{\partial\phi}G^{R(A)}(\epsilon)\to G^{R(A)}(\epsilon)\hat{j}G^{R(A)}(\epsilon). \tag{11}$$

It is important that neither the analytical properties nor frequency is changed in the Green's functions involved.

Thus, the function in the r.h.s. of Eq.(7) takes the form:

$$\begin{aligned}\Re\,\sigma^{(2)}_{eq}(\mathbf{q},\omega)\propto Tr_{\mathbf{k}}\int_{-\infty}^{\infty}\frac{d\epsilon}{4\pi i}\,\Big[&[G^R(\epsilon+\omega)\hat{j}G^R(\epsilon+\omega)G^R(\epsilon)-G^A(\epsilon)\hat{j}G^A(\epsilon)G^A(\epsilon-\omega)+ \\ &+G^R(\epsilon+\omega)G^R(\epsilon)\hat{j}G^R(\epsilon)-G^A(\epsilon)G^A(\epsilon-\omega)\hat{j}G^A(\epsilon-\omega)]\tanh\left(\frac{\epsilon}{2T}\right)+ \\ &+2[G^R(\epsilon+\omega)\hat{j}G^R(\epsilon+\omega)G^A(\epsilon)+G^R(\epsilon+\omega)G^A(\epsilon)\hat{j}G^A(\epsilon)]\,[n_F(\epsilon)-n_F(\epsilon+\omega)]\Big]+(\omega\to-\omega).\end{aligned} \tag{12}$$

Eq.(12) can be also obtained by the direct analytical continuation of the expression $K(i\omega_l,i\omega_m)$ given by the triangle diagram in Fig.1a.:

$$K(i\omega_l,i\omega_m)=Tr_{\mathbf{k}}T\sum_n G(i\epsilon_n)G(i\epsilon_n+i\omega_l)\hat{j}G(i\epsilon_n-i\omega_m). \tag{13}$$

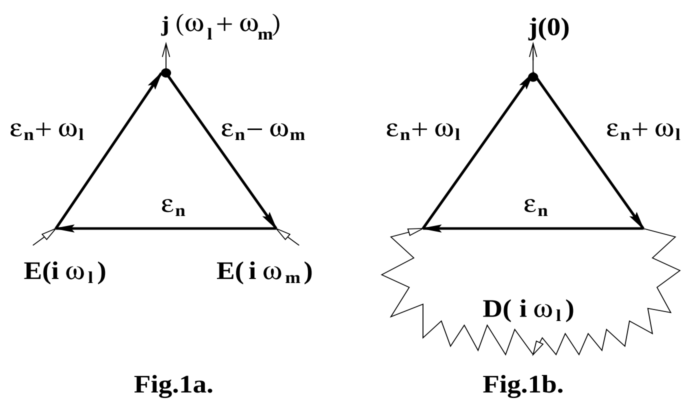

FIG. 1. a). A generic traingle diagram corresponding to Eq.(13). Bold lines denote electron Green's functions with Matsubare frequencies indicated; b).The Fock contribution to an equilibrium persistent current. The field vertices are connected with a photon Green's function.

The equilibrium Matsubara diagrammatic technique (see Fig.1b.) requires $i\omega_l=-i\omega_m$, since the corresponding vertices are connected by the photon Green's function $D(\mathbf{q},i\omega_l)$. That is why in order to obtain Eq.(12) one should *first set* $i\omega_l=-i\omega_m$ in Eq.(13) and *then* perform an analytical continuation of $K(i\omega_l)=K(i\omega_l,-i\omega_l)$ into the upper half-plane of ω. One can check that:

$$\Re\sigma^{(2)}_{eq}(\mathbf{q},\omega)\propto K^R(\omega)+K^R(-\omega)+c.c. \tag{14}$$

The nonlinear conductivity $\sigma^{(2)}(\omega,-\omega)$ can also be obtained from a generic triangle diagram Fig.1a. However, the proper procedure is completely different. It is determined by the *causal* nature of the system response to an external field. For a generic quadratic response to a time-dependent external field $E(t)$:

$$I(t)=\int_{-\infty}^{\infty}d\tau_1\int_{-\infty}^{\infty}d\tau_2\,\sigma(\tau_1,\tau_2)\,E(t-\tau_1)\,E(t-\tau_2). \tag{15}$$

causality requires that $\sigma(\tau_1,\tau_2)\propto\theta(\tau_1)\theta(\tau_2)$, where $\theta(x)$ is a step function. This means that the generic nonlinear conductivity $\sigma(\omega_1,\omega_2)$ is an analytical function in the upper half-plane of *both* ω_1 and ω_2.

This is the key point in obtaining the nonlinear response from the Matsubara diagrammatic technique [7]. One should first find an analytical continuation $K^{(R,R)}(\omega_1,\omega_2)$ of a *generic* triangle diagram $K(i\omega_l,i\omega_m)$ into the upper half-plane of *both* frequencies, and then set $\omega_1=-\omega_2=\omega$ to obtain a DC nonlinear response:

$$\sigma^{(2)}_{kin}(\mathbf{q},\omega,-\omega)=K^{R,R}(\omega,-\omega)+K^{R,R}(-\omega,\omega). \tag{16}$$

By performing the analytical continuation in Eq.(13) we arrive at:

$$\sigma^{(2)}_{kin}(\omega,-\omega)\propto Tr_{\mathbf{k}}\int_{-\infty}^{\infty}\frac{d\epsilon}{4\pi i}\,\Big[[G^R(\epsilon-\omega)G^R(\epsilon)\hat{j}G^R(\epsilon)-G^A(\epsilon-\omega)G^A(\epsilon)\hat{j}G^A(\epsilon)]\tanh\left(\frac{\epsilon}{2T}\right)+$$
$$2[G^A(\epsilon)G^R(\epsilon+\omega)\hat{j}G^A(\epsilon+\omega)-G^R(\epsilon)G^R(\epsilon+\omega)\hat{j}G^A(\epsilon+\omega)][n_F(\epsilon)-n_F(\epsilon+\omega)]\Big]+(\omega\to-\omega). \tag{17}$$

Now it is explicitly seen that the $R-A$ structure of Eqs.(12),(17) is different even before disorder-averaging is done. The quantity $\sigma^{(2)}_{eq}(\mathbf{q},\omega)$ in Eq.(12) has a structure of a derivative of the linear AC polarizability with respect to the DC flux. Therefore the there is no change of analytical properties in passing the current vertex. On the contrary, the nonlinear conductance $\sigma^{(2)}_{kin}(\omega,-\omega)$ is a derivative of the AC conductance with respect to the AC flux. Correspondingly, R is changed to A in passing the current vertex. This proves invalidity of the basic assumption (7) of Kopietz and Völker Ref. [6]. The quantities Eqs.(12),(17) have different physical meaning and the difference in their analytical structure is crucial for the functional dependence of their disorder-average values at high frequencies.

To this end, we note that only such terms in Eqs.(12),(17) that contain both retarded (R) and advanced (A) electron Green's functions make a significant contribution to the disorder-averaged values. This is because of the diffusion propagators (diffusons and cooperons) $P(\mathbf{q},\omega)\propto Tr_{\mathbf{q}}\overline{G^R(\epsilon+\omega)G^A(\epsilon)}$ which are singular at small q and ω. It implies that only last lines in Eqs.(12),(17) are relevant. Because R is changed to A and frequency is unchanged in passing the DC current vertex in Eq.(17), it is possible to build a diffusion propagator (cooperon) of zero frequency in the kinetic case. There is no such a possibility in the equilibrium case, since the R-A structure is unchanged in passing the DC current vertex. The relevant two-cooperon diagrams are shown in Fig.2.

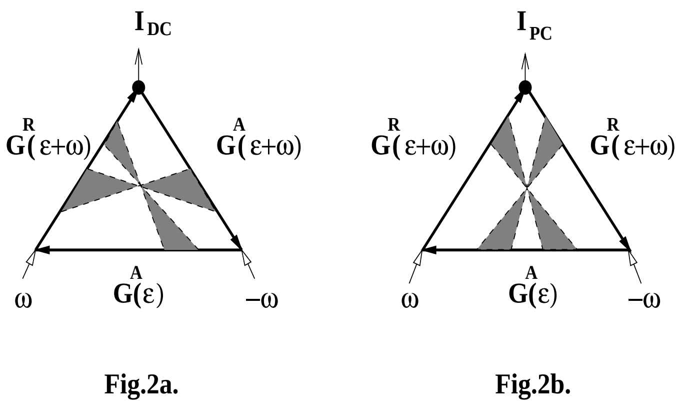

FIG. 2. a). The leading contribution to the disorder-average kinetic coefficient $\sigma^{(2)}_{kin}(\omega,-\omega)$; b). The leading contribution to the disorder-average value of $\sigma^{(2)}_{eq}(0,\omega)$. The bold lines denote electron Green's functions; the shadowed area between two dotted lines denotes the fan series (cooperon) $P(\mathbf{q},\omega)$ with the frequency ω equal to the difference of frequencies of electron Green's functions which are connected by the dotted lines.

It turns out that both terms in the last lines of Eqs.(12),(17) lead to identical contributions to $\sigma^{(2)}_{kin}(\omega,-\omega)$ and $\Re\sigma^{(2)}_{eq}(0,\omega)$, respectively. In the case of nonlinear response where one of the cooperons is of zero frequency, the summation over the cooperon momenta is mainly contributed by small momenta $|k|\sim 1/L\ll 1/L_{\omega}=\sqrt{\omega/D}$. Then one can set $k=0$ in the second cooperon and obtain a power-law ω-dependence. In the equilibrium case both cooperons are of the same frequency ω, since they appear due to differentiation with respect to the DC flux of a single-cooperon diagram for the Aharonov-Bohm effect in the linear AC polarizability. As a result, the diagram turns out to be exponentially small in full agreement with the exponentially small Aharonov-Bohm oscillations of the polarizability at high frequencies.

In conclusion, we have shown that the analytical structure of the expression for the *free-electron nonlinear* conductance $\sigma^{(2)}_{kin}(\mathbf{q},\omega,-\omega)$ is different from that of the coefficient $\sigma^{(2)}_{eq}(\mathbf{q};\omega)$ that couples the Fock contribution to the equilibrium persistent current and the spectral density of the electric field fluctuations in the system of *interacting* electrons. The difference is caused by the *causality* requirement that determines the nonlinear response to an external field but is irrelevant for the interaction contribution to equilibrium quantities. As a result, at high frequencies

$\omega \gg E_c$ the free-electron nonlinear conductance $\sigma^{(2)}_{kin}(\omega,-\omega) \propto 1/\omega^2$, while $\sigma^{(2)}_{eq}(\mathbf{q} \to 0;\omega)$ is exponentially small. This analysis warns against a possible mistake when one applies methods developed for describing kinetic phenomena to equilibrium systems and *vice versa*.

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